Spin tunneling properties in mesoscopic magnets: effects of a magnetic field

Rong Lü,* Hui Pan, Jia-Lin Zhu, and Bing-Lin Gu

Center for Advanced Study, Tsinghua University, Beijing 100084, P. R. China

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Abstract

The tunneling of a giant spin at excited levels is studied theoretically in mesoscopic magnets with a magnetic field at an arbitrary angle in the easy plane. Different structures of the tunneling barriers can be generated by the magnetocrystalline anisotropy, the magnitude and the orientation of the field. By calculating the nonvacuum instanton solution explicitly, we obtain the tunnel splittings and the tunneling rates for different angle ranges of the external magnetic field ($\theta_H = \pi/2$ and $\pi/2 < \theta_H < \pi$). The temperature dependences of the decay rates are clearly shown for each case. It is found that the tunneling rate and the crossover temperature depend on the orientation of the external magnetic field. This feature can be tested with the use of existing experimental techniques.

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*Author to whom the correspondence should be addressed.

Electronic address: rlu@castu.tsinghua.edu.cn

I. INTRODUCTION

Recently, nanospin systems have emerged as good candidates to display quantum phenomena at a mesoscopic or macroscopic scale.[1] Theoretical investigations showed that quantum tunneling was possible in ferromagnetic (FM) nanoparticles containing as much as $10^5 - 10^6$ spins.[1] At extremely low temperature, the magnitude of the total magnetization $\mathbf{M}$ is frozen out and thereby its direction becomes the only dynamical variable. In the absence of an external magnetic field, the magnetocrystalline anisotropy can create energetically degenerate easy directions depending on the crystal symmetry. Tunneling between neighboring states removes the degeneracy of the original ground states and leads to a level splitting. This phenomenon is called macroscopic quantum coherence (MQC). However, MQC is hard to be observed in experiments without controlling the height and the width of the barrier. It has been believed that a magnetic field is a good external parameter to make the quantum tunneling observable. By applying a magnetic field in a proper direction, one of the two energetically equivalent orientations becomes metastable and the magnetization vector can escape from the metastable state through the barrier to a stable one, which is called macroscopic quantum tunneling (MQT). A large number of experiments involving resonance measurements, magnetic relaxation, and hysteresis loop study, Mössbauer spectroscopy, and neutron scattering study for various systems showed either temperature-independent relaxation phenomena or a well-defined resonance depending exponentially on the number of total spins, which supported the idea of magnetic quantum tunneling.[1]

To our knowledge, the tunneling of a single spin degree of freedom was first studied by Korenblit and Shender in 1978.[2] More recently, the tunneling problem of the magnetization reversal was studied extensively for the single-domain FM nanoparticles in a magnetic field applied at an arbitrary angle. This problem was studied by Zaslavskii with the help of mapping the spin system onto a one-dimensional particle system.[3] For the same system, Miguel and Chudnovsky[4] calculated the tunneling rate by applying the imaginary-time path integral, and demonstrated that the angular and field dependences of the tunneling exponent

obtained by Zaslavskii's method and by the path-integral method coincide precisely. Kim and Hwang performed a calculation based on the instanton technique for FM particles with biaxial and tetragonal crystal symmetry,[5] and Kim extended the tunneling rate for biaxial crystal symmetry to a finite temperature.[6] The quantum-classical transition of the escape rate for uniaxial spin system in an arbitrarily directed field was investigated by Garanin, Hidalgo and Chudnovsky with the help of mapping onto a particle moving in a double-well potential.[7] The switching field measurement was carried out on single-domain FM nanoparticles of Barium ferrite (BaFeCoTiO) containing about $10^5 - 10^6$ spins.[8] The measured angular dependance of the crossover temperature was found to be in excellent agreement with the theoretical prediction,[4] which strongly suggests the quantum tunneling of magnetization in the BaFeCoTiO nanoparticles. Lü *et al.* studied the quantum tunneling of the Néel vector in single-domain antiferromagnetic (AFM) nanoparticles with biaxial, tetragonal, and hexagonal crystal symmetry in an arbitrarily directed field.[9]

It is noted that the previous results of spin tunneling at excited levels in an arbitrarily directed field were obtained by numerically solving the equation of motion satisfied by the least trajectory,[6] and the system considered in Ref. 6 had the simple biaxial crystal symmetry. The purpose of this paper is to present an analytical study of the quantum tunneling *at excited levels* in the FM particles with an arbitrarily directed field. Moreover, the system considered in this paper has a much more complex structure (i. e., *the general structure in experiments*), such as trigonal, tetragonal, and hexagonal crystal symmetry. By applying an arbitrarily directed magnetic field, the problem does not possess any symmetry and for that reason is more difficult mathematically. However, it is worth pursuing because of its significance for experiments and the easiest to implement in practice. Since the result of spin tunneling at excited levels for tetragonal symmetry is a generalization of that of tunneling at ground-state levels studied by Kim and Hwang,[5] we can compare our results with theirs by taking the low-energy limit. We will show that MQC and MQT can be consecutively observed by changing the direction of magnetic field, and discuss their dependence on the direction and the magnitude of field. The dependence of the crossover temperature

T_c and the magnetic viscosity (which is the inverse of WKB exponent at the quantum-tunneling-dominated regime $T \ll T_c$) on the direction and the magnitude of the field, and the magnetic anisotropies is expected to be observed in future experiments on individual single-domain particles with different magnetocrystalline anisotropies. Both the nonvacuum (or thermal) instanton or bounce solution, the WKB exponents and the preexponential factors are evaluated exactly for different angle ranges of the magnetic field ($\theta_H = \pi/2$ and $\pi/2 < \theta_H < \pi$). The low-energy limit of our results agrees well with that of ground-state spin tunneling. In order to compare theories with experiments, predictions of the crossover temperature corresponding to the transition from classical to quantum behavior and the temperature dependence of the decay rate are clearly shown in this paper. Both variables are expressed as a function of parameters which can be changed experimentally, such as the number of total spins, the effective anisotropy constants, the strength and orientation of applied magnetic field. Our results show that the distinct angular dependence, together with the dependence of the WKB tunneling rate on the strength of the external magnetic field, may provide an independent experimental test for the spin tunneling at excited levels in nanoscale magnets. When the effective magnetic anisotropy of the particle is known, our theoretical results give clear predictions with no fitting parameters. Therefore, quantum spin tunneling could be studied as a function of the effective magnetic anisotropy. Our results should be helpful for future experiments on spin tunneling in single-domain particles with different magnetocrystalline anisotropies.

This paper is structured in the following way. In Sec. II, we review briefly some basic ideas of spin tunneling in FM particles. And we discuss the fundamentals concerning the computation of level splittings and tunneling rates of excited states in the double-well-like potential. In Secs. III, IV, and V, we study the spin tunneling at excited levels for FM particles with trigonal, tetragonal and hexagonal crystal symmetry in an external magnetic field applied in the ZX plane with a range of angles $\pi/2 \leq \theta_H < \pi$, respectively. The conclusions are presented in Sec. VI.

II. PHYSICAL MODEL OF SPIN TUNNELING IN FM PARTICLES

For a spin tunneling problem, the tunnel splitting or the tunneling rate is determined by the imaginary-time transition amplitude from an initial state $|i\rangle$ to a final state $|f\rangle$ as

$$\mathcal{U}_{fi} = \langle f | e^{-\mathcal{H}T} |i\rangle = \int \mathcal{D}\Omega \exp\left(-\mathcal{S}_E\right), \tag{1}$$

where $\mathcal{S}_E$ is the Euclidean action and $\mathcal{D}\Omega$ is the measure of the path integral. In the spin-coherent-state representation, the Euclidean action is

$$\mathcal{S}_E\left(\theta,\phi\right) = \frac{V}{\hbar}\int d\tau \left[i\frac{M_0}{\gamma}\left(\frac{d\phi}{d\tau}\right) - i\frac{M_0}{\gamma}\left(\frac{d\phi}{d\tau}\right)\cos\theta + E\left(\theta,\phi\right)\right], \tag{2}$$

where V is the volume of the FM particle and γ is the gyromagnetic ratio. $M_0 = |\mathbf{M}| = \hbar\gamma S/V$, where S is the total spin of FM particles. It is noted that the first two terms in Eq. (2) define the Berry phase or Wess-Zumino, Chern-Simons term which arises from the nonorthogonality of spin coherent states in the north-pole parametrization. The Wess-Zumino term has a simple topological interpretation. For a closed path, this term equals $-iS$ times the area swept out on the unit sphere between the path and the north pole. The first term in Eq. (2) is a total imaginary-time derivative, which has no effect on the classical equations of motion, but it is crucial for the spin-parity effects.[1,10–14] However, for the closed instanton or bounce trajectory described in this paper (as shown in the following), this time derivative gives a zero contribution to the path integral, and therefore can be omitted.

In the semiclassical limit, the dominant contribution to the transition amplitude comes from finite action solution (instanton or bounce) of the classical equation of motion. The instanton's contribution to the tunneling rate Γ or the tunnel splitting Δ is given by[1]

$$\Gamma \ (\text{or} \ \ \Delta) = A\omega_p \left(\frac{\mathcal{S}_{cl}}{2\pi}\right)^{1/2} e^{-\mathcal{S}_{cl}}, \tag{3}$$

where ω_p is the oscillation frequency in the well, $\mathcal{S}_{cl}$ is the classical action, and the prefactor A originates from the quantum fluctuations about the classical path. It is noted that Eq. (3) is based on quantum tunneling at the level of ground state, and the temperature dependence of the tunneling rate (i.e., tunneling at excited levels) is not taken into account.

However, the instanton technique is suitable only for the evaluation of the tunneling rate or the tunnel splitting at the vacuum level, since the usual (vacuum) instantons satisfy the vacuum boundary conditions. In this paper, we will calculate the nonvacuum instantons corresponding to quantum tunneling at excited levels.

For a particle moving in a double-well-like potential $U(x)$, the level splittings of degenerate excited levels or the imaginary parts of the metastable levels at an energy $E > 0$ are given by the following formula in the WKB approximation,[16]

$$\Delta E \ \ (\text{or } \ \mathrm{Im}\, E) = \frac{\omega(E)}{\pi} \exp\left[-\mathcal{S}(E)\right], \tag{4}$$

and the imaginary-time action is

$$\mathcal{S}(E) = 2\sqrt{2m} \int_{x_1(E)}^{x_2(E)} dx \sqrt{U(x) - E}, \tag{5}$$

where $x_{1,2}(E)$ are the turning points for the particle oscillating inside the inverted potential $-U(x)$. $\omega(E) = 2\pi/t(E)$ is the energy-dependent frequency, and $t(E)$ is the period of the real-time oscillation in the potential well,

$$t(E) = \sqrt{2m} \int_{x_3(E)}^{x_4(E)} \frac{dx}{\sqrt{E - U(x)}}, \tag{6}$$

where $x_{3,4}(E)$ are the turning points for the particle oscillating inside the potential $U(x)$.

III. MQC AND MQT FOR TRIGONAL CRYSTAL SYMMETRY

In this section, we study the quantum tunneling of the magnetization vector in single-domain FM nanoparticles with trigonal crystal symmetry. The external magnetic field is applied in the ZX plane, at an angle in the range of $\pi/2 \leq \theta_H < \pi$. Now the total energy $E(\theta, \phi)$ can be written as

$$E(\theta, \phi) = K_1 \sin^2\theta - K_2 \sin^3\theta \cos(3\phi) - M_0 H_x \sin\theta \cos\phi - M_0 H_z \cos\theta + E_0, \tag{7}$$

where K_1 and K_2 are the magnetic anisotropy constants satisfying $K_1 \gg K_2 > 0$, and E_0 is a constant which makes $E(\theta, \phi)$ zero at the initial orientation. As the magnetic field is

applied in the ZX plane, $H_x = H\sin\theta_H$ and $H_z = H\cos\theta_H$, where H is the magnitude of the field and θ_H is the angle between the magnetic field and the $\widehat{z}$ axis.

By introducing the dimensionless parameters as

$$\overline{K}_2 = K_2/2K_1, \overline{H}_x = H_x/H_0, \overline{H}_z = H_z/H_0, \tag{8}$$

Eq. (7) can be rewritten as

$$\overline{E}(\theta,\phi) = \frac{1}{2}\sin^2\theta - \overline{K}_2\sin^3\theta\cos(3\phi) - \overline{H}_x\sin\theta\cos\phi - \overline{H}_z\cos\theta + \overline{E}_0, \tag{9}$$

where $E(\theta,\phi) = 2K_1\overline{E}(\theta,\phi)$, and $H_0 = 2K_1/M_0$. At finite magnetic field, the plane given by $\phi = 0$ is the easy plane, on which $\overline{E}(\theta,\phi)$ reduces to

$$\overline{E}(\theta,\phi=0) = \frac{1}{2}\sin^2\theta - \overline{K}_2\sin^3\theta - \overline{H}\cos(\theta-\theta_H) + \overline{E}_0. \tag{10}$$

We denote θ_0 to be the initial angle and θ_c the critical angle at which the energy barrier vanishes when the external magnetic field is close to the critical value $\overline{H}_c(\theta_H)$ (to be calculated in the following). Then, the initial angle θ_0 satisfies $\left[d\overline{E}(\theta,\phi=0)/d\theta\right]_{\theta=\theta_0} = 0$, the critical angle θ_c and the dimensionless critical field $\overline{H}_c$ satisfy both $\left[d\overline{E}(\theta,\phi=0)/d\theta\right]_{\theta=\theta_c,\overline{H}=\overline{H}_c} = 0$ and $\left[d^2\overline{E}(\theta,\phi=0)/d\theta^2\right]_{\theta=\theta_c,\overline{H}=\overline{H}_c} = 0$. After some algebra, $\overline{H}_c(\theta_H)$ and θ_c are found to be

$$\overline{H}_c = \frac{1}{\left[(\sin\theta_H)^{2/3} + |\cos\theta_H|^{2/3}\right]^{3/2}}\left[1 - 3\overline{K}_2\frac{1}{\left(1+|\cot\theta_H|^{2/3}\right)^{1/2}}\right.$$
$$\left. + 6\overline{K}_2\frac{1}{\left(1+|\cot\theta_H|^{2/3}\right)^{3/2}}\right], \tag{11a}$$

$$\sin^2\theta_c = \frac{1}{1+|\cot\theta_H|^{2/3}}\left[1 - 2\overline{K}_2\frac{|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^{1/2}} - 4\overline{K}_2\frac{|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^{3/2}}\right]. \tag{11b}$$

Now we consider the limiting case that the external magnetic field is slightly lower than the critical field, i.e., $\epsilon = 1-\overline{H}/\overline{H}_c \ll 1$. At this practically interesting situation, the barrier height is low and the width is narrow, and therefore the tunneling rate in MQT or the tunnel splitting in MQC is large. Introducing $\eta \equiv \theta_c - \theta_0$ ($|\eta| \ll 1$ in the limit of $\epsilon \ll 1$), expanding

$\left[d\overline{E}\left(\theta,\phi=0\right)/d\theta\right]_{\theta=\theta_0}=0$ about θ_c, and using the relations $\left[d\overline{E}\left(\theta,\phi=0\right)/d\theta\right]_{\theta=\theta_c,\overline{H}=\overline{H}_c}=0$ and $\left[d^2\overline{E}\left(\theta,\phi=0\right)/d\theta^2\right]_{\theta=\theta_c,\overline{H}=\overline{H}_c}=0$, we obtain the approximation equation for η in the order of $\epsilon^{3/2}$,

$$\begin{aligned}&-\epsilon\overline{H}_c\sin\left(\theta_c-\theta_H\right)-\eta^2\left(\frac{3}{4}\sin 2\theta_c+3\overline{K}_2\cos 3\theta_c\right)\\&+\eta\left[\epsilon\overline{H}_c\cos\left(\theta_c-\theta_H\right)+\eta^2\left(\frac{1}{2}\cos 2\theta_c-3\overline{K}_2\sin 3\theta_c\right)\right]=0.\end{aligned}\tag{12}$$

Then $\overline{E}\left(\theta,\phi\right)$ reduces to the following equation in the limit of small ϵ,

$$\overline{E}\left(\delta,\phi\right)=2\overline{K}_2\sin^2\left(3\phi/2\right)\sin^3\left(\theta_0+\delta\right)+\overline{H}_x\sin\left(\theta_0+\delta\right)\left(1-\cos\phi\right)+\overline{E}_1\left(\delta\right),\tag{13}$$

where $\delta\equiv\theta-\theta_0$ ($|\delta|\ll 1$ in the limit of $\epsilon\ll 1$), and $\overline{E}_1\left(\delta\right)$ is a function of only δ given by

$$\begin{aligned}\overline{E}_1\left(\delta\right)&=-\frac{1}{2}\left[\overline{H}_c\sin\left(\theta_c-\theta_H\right)-\overline{K}_2\left(\cos^3\theta_c-\frac{3}{2}\sin^2\theta_c\cos\theta_c\right)\right]\left(3\delta^2\eta-\delta^3\right)\\&-\frac{1}{2}\left[\overline{H}_c\cos\left(\theta_c-\theta_H\right)-3\overline{K}_2\left(\sin^3\theta_c-4\sin\theta_c\cos^2\theta_c\right)\right]\left[\delta^2\left(\epsilon-\frac{3}{2}\eta^2\right)+\delta^3\eta-\frac{1}{4}\delta^4\right]\\&-\frac{3}{2}\overline{K}_2\left(\sin^3\theta_c-4\sin\theta_c\cos^2\theta_c\right)\delta^2\epsilon.\end{aligned}\tag{14}$$

In the following, we will investigate the tunneling behaviors of the magnetization vector at excited levels in FM particles with trigonal crystal symmetry at different angle ranges of the external magnetic field as $\theta_H=\pi/2$ and $\pi/2<\theta_H<\pi$, respectively.

A. $\theta_H=\pi/2$

For $\theta_H=\pi/2$, we have $\theta_c=\pi/2$ from Eq. (11b) and $\eta=\sqrt{2\epsilon}\left(1+\frac{9}{2}\overline{K}_2\right)$ from Eq. (12). Eqs (13) and (14) show that ϕ is very small for the full range of angles $\pi/2\leq\theta_H<\pi$. Performing the Gaussian integration over ϕ, we can map the spin system onto a particle moving problem in the one-dimensional potential well. Now the imaginary-time transition amplitude Eqs. (1) and (2) becomes

$$\begin{aligned}\mathcal{U}_{fi}&=\int d\delta\exp\left(-\mathcal{S}_E\left[\delta\right]\right),\\&=\int d\delta\exp\left\{-\int d\tau\left[\frac{1}{2}m\left(\frac{d\delta}{d\tau}\right)^2+U\left(\delta\right)\right]\right\},\end{aligned}\tag{15}$$

with the effective mass

$$m = \frac{\hbar S^2}{2VK_1\left[1-\epsilon+9\overline{K}_2\right]},$$

and the effective potential

$$U(\delta) = \frac{K_1 V}{4\hbar}\delta^2(\delta-2\eta)^2. \tag{16}$$

The plot of the effective potential $\overline{E}_1(\delta)$ as a function of $\delta\,(=\theta-\theta_0)$ for $\theta_H=\pi/2$ is shown in Fig. 1, and $\hbar U(\delta) = 2K_1 V\overline{E}_1(\delta)$. The problem is one of MQC, where the magnetization vector resonates coherently between the energetically degenerate easy directions at $\delta = 0$ and $\delta = 2\sqrt{2\epsilon}\left(1+\frac{9}{2}\overline{K}_2\right)$ separated by a classically impenetrable barrier at $\delta = \sqrt{2\epsilon}\left(1+\frac{9}{2}\overline{K}_2\right)$.

The nonvacuum (or thermal) instanton configuration δ_p which minimizes the Euclidean action in Eq. (16) satisfies the equation of motion

$$\frac{1}{2}m\left(\frac{d\delta_p}{d\tau}\right)^2 - U(\delta_p) = -E, \tag{17}$$

where $E>0$ is a constant of integration, which can be viewed as the classical energy of the pseudoparticle configuration. Then the kink-solution is

$$\delta_p = \eta + \sqrt{\eta^2-\alpha}\,\mathrm{sn}(\omega_1\tau, k), \tag{18}$$

where $\alpha = 2\sqrt{\frac{\hbar E}{K_1 V}}$, and $\omega_1 = \sqrt{\frac{K_1 V}{2\hbar m}}\sqrt{\eta^2+\alpha}$. $\mathrm{sn}(\omega_1\tau, k)$ is the Jacobian elliptic sine function of modulus $k = \sqrt{\frac{\eta^2-\alpha}{\eta^2+\alpha}}$. The Euclidean action of the nonvacuum instanton configuration Eq. (18) over the domain $(-\beta,\beta)$ is found to be

$$\mathcal{S}_p = \int_{-\beta}^{\beta} d\tau\left[\frac{1}{2}m\left(\frac{d\delta_p}{d\tau}\right)^2 + U(\delta_p)\right] = W + 2E\beta, \tag{19a}$$

with

$$W = \frac{8}{3}\sqrt{\frac{K_1 V m}{\hbar}}\left(1+\frac{27}{2}\overline{K}_2\right)\epsilon^{3/2}\frac{1}{\sqrt{1-k'^2/2}}\left[E(k) - \frac{k'^2}{2-k'^2}K(k)\right], \tag{19b}$$

where $k'^2 = 1-k^2$, and $\beta = 1/k_B T$ with k_B the Boltzmann constant. $K(k)$ and $E(k)$ are the complete elliptic integral of the first and second kind, respectively. The general formula

Eq. (4) gives the tunnel splittings of excited levels as $\Delta E = \frac{\omega(E)}{\pi}\exp(-W)$, where W is shown in Eq. (19b), and $\omega(E) = \frac{2\pi}{t(E)}$ is the energy-dependent frequency. For this case, the period $t(E)$ is found to be

$$t(E) = \sqrt{2m}\int_{\delta_1}^{\delta_2}\frac{d\delta}{\sqrt{E - U(\delta)}} = 2\sqrt{\frac{2\hbar m}{K_1 V}}\frac{1}{\sqrt{\eta^2+\alpha}}K(k'), \tag{20}$$

where $\delta_1 = \eta + \sqrt{\eta^2-\alpha}$, and $\delta_2 = \eta + \sqrt{\eta^2+\alpha}$. Now we discuss the low energy limit where E is much less than the barrier height. In this case, $k'^4 = \frac{16\hbar E}{K_1 V\eta^4} \ll 1$, so we can perform the expansions of $K(k)$ and $E(k)$ in Eq. (19b) to include terms like k'^4 and $k'^4\ln\left(\frac{4}{k'}\right)$,

$$\begin{aligned} E(k) &= 1 + \frac{1}{2}\left[\ln\left(\frac{4}{k'}\right) - \frac{1}{2}\right]k'^2 + \frac{3}{16}\left[\ln\left(\frac{4}{k'}\right) - \frac{13}{12}\right]k'^4\cdots, \\ K(k) &= \ln\left(\frac{4}{k'}\right) + \frac{1}{4}\left[\ln\left(\frac{4}{k'}\right) - 1\right]k'^2 + \frac{9}{64}\left[\ln\left(\frac{4}{k'}\right) - \frac{7}{6}\right]k'^4\cdots. \end{aligned}$$

With the help of small oscillator approximation for energy near the bottom of the potential well, $E_n = \left(n+\frac{1}{2}\right)\Omega_1$, $\Omega_1 = \sqrt{\frac{1}{m}U''(\delta=0)} = \eta\sqrt{\frac{2K_1 V}{\hbar m}}$, Eq. (19b) is expanded as

$$W = W_0 - \left(n+\frac{1}{2}\right) + \left(n+\frac{1}{2}\right)\ln\left[\frac{1-\frac{\epsilon}{2}-\frac{15}{2}\overline{K}_2}{2^{9/2}S\epsilon^{3/2}}\left(n+\frac{1}{2}\right)\right], \tag{21a}$$

where

$$W_0 = \frac{2^{5/2}}{3}S\epsilon^{3/2}\left(1+\frac{\epsilon}{2}+\frac{15}{2}\overline{K}_2\right). \tag{21b}$$

Then the low-lying energy shift of n-th excited states for FM particles with trigonal crystal symmetry in the presence of an external magnetic field applied perpendicular to the anisotropy axis ($\theta_H = \pi/2$) is

$$\hbar\Delta E_n = \frac{2}{n!\sqrt{\pi}}(K_1 V)\epsilon^{1/2}S^{-1}\left(1-\frac{\epsilon}{2}+\frac{21}{2}\overline{K}_2\right)\left(\frac{2^{9/2}S\epsilon^{3/2}}{1-\frac{\epsilon}{2}-\frac{15}{2}\overline{K}_2}\right)^{n+1/2}\exp(-W_0). \tag{22}$$

For $n=0$, the energy shift of the ground state is

$$\hbar\Delta E_0 = \frac{2^{13/4}}{\sqrt{\pi}}(K_1 V)\epsilon^{5/4}S^{-1/2}\left(1-\frac{\epsilon}{4}+\frac{57}{4}\overline{K}_2\right)\exp(-W_0). \tag{23}$$

Then Eq. (22) can be written as

$$\hbar\Delta E_n = \frac{q_1^n}{n!}\left(\hbar\Delta E_0\right), \tag{24a}$$

where

$$q_1 = \frac{2^{9/2} S \epsilon^{3/2}}{1 - \frac{\epsilon}{2} - \frac{15}{2}\overline{K}_2}. \tag{24b}$$

Since we have obtained the tunnel splittings at excited levels, it is reasonable to study the temperature dependence of the tunneling rate. It is noted that Eqs. (24a) and (24b) are obtained under the condition that the levels in the two wells are degenerate. In more general cases, the transition amplitude between two levels separated by the barrier or the decay rate should be sensitive to this resonance condition for the two levels. If in the case of the potential with two degenerate levels only one of the levels is considered as a perturbative metastable state; however, a fictitious imaginary energy can be calculated by consideration of possible back and forth tunneling (i.e., by regarding the instanton-antiinstanton pair as a bounce-like configuration) in the barrier. Therefore there exists a relation between the level splitting and this imaginary part of metastable energy level, and has been referred to as the Bogomolny-Fateyev relation based on equilibrium thermodynamics[17]

$$\operatorname{Im} E_n = \pi\left(\Delta E_n\right)^2 / 4\omega\left(E_n\right), \tag{25}$$

where $\omega\left(E_n\right)$ is the frequency of oscillations at energy level E_n. At finite temperature T the decay rate $\Gamma = 2\operatorname{Im} E_n$ can be easily found by averaging over the Boltzmann distribution

$$\Gamma\left(T\right) = \frac{2}{Z_0}\sum_n \operatorname{Im} E_n \exp\left(-\hbar E_n \beta\right), \tag{26}$$

where $Z_0 = \sum_n \exp\left(-\hbar E_n \beta\right)$ is the partition function with the harmonic oscillator approximated eigenvalues $E_n = \left(n + \frac{1}{2}\right)\Omega_1$. With the help of the Bogomolny-Fateyev relation Eq. (25), the final result of the tunneling rate at a finite temperature T is found to be

$$\Gamma\left(T\right) = \frac{\pi}{2\Omega_1}\left(1 - e^{-\hbar\Omega_1\beta}\right)\left(\Delta E_0\right)^2 I_0\left(2q_1 e^{-\hbar\Omega_1\beta/2}\right), \tag{27}$$

where ΔE_0 and q_1 are shown in Eqs. (23) and (24b). $I_0\left(x\right) = \sum_{n=0}\left(x/2\right)^{2n}/\left(n!\right)^2$ is the modified Bessel function.

Now we discuss briefly the dissipation effect on spin tunneling. For a spin tunneling problem, it is important to consider the discrete level structure. It was quantitatively shown that the phenomenon of MQC depends crucially on the width of the excited levels in the right well.[18] Including the effects of dissipation, the decay rate, in particular, is given by[18–20]

$$\Gamma_n = \frac{1}{2}\left(\Delta E_n\right)^2 \sum_{n'} \frac{\Omega_{nn'}}{\left(E_n - E_{n'}\right)^2 + \Omega_{nn'}^2}, \tag{28}$$

where ΔE_n is the level splitting, n' are the levels in the other well and $\Omega_{nn'}$ is the sum of the linewidths of the nth and n'th levels caused by the coupling of the system to the environment. For the exact resonance conditions, the temperature dependence of the decay rate is

$$\Gamma\left(T\right) = \sum_{n} \frac{\left(\Delta E_n\right)^2}{2\Omega_n} \exp\left(-\hbar E_n \beta\right), \tag{29}$$

where the level broadening Ω_n contains all the details of the coupling between the magnet and its environment. If the width caused by the dissipative coupling sufficiently large, the levels overlap, so that the problem is more or less equivalent to the tunneling into the structureless continuum. In this case, the results obtained in this paper should be changed by including the dissipation. It is noted that the purpose of this paper is to study the spin tunneling at excited levels for single-domain FM particles in magnetic field at sufficiently low temperatures. Strong dissipation is hardly the case for single-domain magnetic particles,[21] and thereby our results are expected to hold. It has been argued that the decay rate should oscillate on the applied magnetic field depending on the relative magnitude between the width and the level spacing.[12,13,18,20,22] However, it is not clear, to our knowledge, what should be the effect of finite temperature in the problem of spin tunneling. The full analysis of spin tunneling onto the precession levels remains an open problem.

B. $\pi/2 < \theta_H < \pi$

For $\pi/2 < \theta_H < \pi$, the critical angle θ_c is in the range of $0 < \theta_c < \pi/2$, and $\eta \approx \sqrt{2\epsilon/3}$. By applying the similar method, the problem can be mapped onto a problem of

one-dimensional motion by integrating out ϕ, and for this case the effective mass m and the effective potential $U(\delta)$ in Eq. (15) are found to be

$$m = \frac{\hbar S^2\left(1+|\cot\theta_H|^{2/3}\right)}{2K_1V\left[1-\epsilon+9\overline{K}_2\left(1+|\cot\theta_H|^{2/3}\right)^{1/2}-\overline{K}_2\frac{3-|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^{1/2}}+2\overline{K}_2\frac{3+|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^{3/2}}\right]}, \tag{30a}$$

and $U(\delta)=3U_0q^2\left(1-\frac{2}{3}q\right)$, with $q=3\delta/2\sqrt{6\epsilon}$, and

$$U_0=\frac{2^{7/2}}{3^{3/2}}\frac{K_1V}{\hbar}\epsilon^{3/2}\frac{|\cot\theta_H|^{1/3}}{1+|\cot\theta_H|^{2/3}}\left[1-\frac{15}{2}\overline{K}_2\frac{1}{\left(1+|\cot\theta_H|^{2/3}\right)^{1/2}}\right]. \tag{30b}$$

The dependence of the effective potential $\overline{E}_1(\delta)$ on $\delta\,(=\theta-\theta_0)$ for $\theta_H=3\pi/4$ is plotted in Fig. 2, and $\hbar U(\delta)=2K_1V\overline{E}_1(\delta)$. The problem now becomes one of MQT, where the magnetization vector escapes from the metastable state at $\delta=0$ through the barrier by quantum tunneling.

The nonvacuum bounce configuration with an energy $E>0$ is found to be

$$\delta_p=\frac{2}{3}\sqrt{6\epsilon}\left[a-(a-b)\,\mathrm{sn}^2(\omega_2\tau,k)\right], \tag{31}$$

where

$$\omega_2=\frac{1}{2}\sqrt{\frac{3U_0}{2m\epsilon}}\sqrt{a-c}. \tag{32}$$

$a(E)>b(E)>c(E)$ denote three roots of the cubic equation

$$q^3-\frac{3}{2}q^2+\frac{E}{2U_0}=0. \tag{33}$$

$\mathrm{sn}(\omega_2\tau,k)$ is the Jacobian elliptic sine function of modulus $k=\sqrt{\frac{a-b}{a-c}}$.

The classical action of the nonvacuum bounce configuration Eq. (31) is

$$\mathcal{S}_p=\int_{-\beta}^{\beta}d\tau\left[\frac{1}{2}m\left(\frac{d\delta_p}{d\tau}\right)^2+U(\delta_p)\right]=W+2E\beta, \tag{34a}$$

with

$$W=\frac{2^{9/2}}{5\times 3^{3/2}}\sqrt{m\epsilon U_0}\,(a-c)^{5/2}\left[2\left(k^4-k^2+1\right)E\left(k\right)-\left(1-k^2\right)\left(2-k^2\right)K\left(k\right)\right]. \quad (34b)$$

The period $t\left(E\right)$ of this case is found to be

$$t\left(E\right)=\sqrt{2m}\int_c^b\frac{d\delta}{\sqrt{E-U\left(\delta\right)}}=4\sqrt{\frac{2\epsilon m}{3U_0\left(a-c\right)}}K\left(k'\right), \quad (35)$$

where $k'^2=1-k^2$. Then the general formula Eq. (4) gives the imaginary parts of the metastable energy levels as $\operatorname{Im}E=\frac{\omega(E)}{\pi}\exp\left(-W\right)$, where $\omega\left(E\right)=\frac{2\pi}{t(E)}$, and W is shown in Eq. (34b).

Here we discuss the low energy limit of the imaginary part of the metastable energy levels. For this case, $E_n=\left(n+\frac{1}{2}\right)\Omega_2$, $\Omega_2=\sqrt{\frac{1}{m}U''\left(\delta=0\right)}=\frac{3}{2}\sqrt{\frac{U_0}{m\epsilon}}$, $a\approx\frac{3}{2}\left(1-\frac{k'^2}{4}\right)$, $b\approx\left(\frac{3}{4}k'^2\right)\left(1+\frac{3}{4}k'^2\right)$, $c\approx-\frac{3}{4}k'^2\left(1+\frac{1}{4}k'^2\right)$, and $k'^4=\frac{16E}{27U_0}\ll 1$. After some calculations, we obtain the imaginary part of the low-lying metastable excited levels as $\hbar\operatorname{Im}E_n=\frac{q_2^n}{n!}\left(\hbar\operatorname{Im}E_0\right)$, where

$$q_2=\frac{2^{25/4}\times 3^{5/4}S\epsilon^{5/4}\left|\cot\theta_H\right|^{1/6}}{1-\frac{\epsilon}{2}+\frac{9}{2}\overline{K}_2\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{1/2}+\frac{1}{4}\overline{K}_2\frac{2+9\left|\cot\theta_H\right|^{2/3}}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{1/2}}+\overline{K}_2\frac{3+\left|\cot\theta_H\right|^{2/3}}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{3/2}}}.$$

The imaginary part of the metastable ground-state level is

$$\hbar\operatorname{Im}E_0=\frac{3^{13/9}\times 2^{31/8}}{\sqrt{\pi}}\left(K_1V\right)\epsilon^{7/8}S^{-1/2}\frac{\left|\cot\theta_H\right|^{1/4}}{1+\left|\cot\theta_H\right|^{2/3}}\left[1-\frac{\epsilon}{4}+\frac{9}{4}\overline{K}_2\left(1+\left|\cot\theta_H\right|^{2/3}\right)\right.$$
$$\left.-\frac{1}{8}\overline{K}_2\frac{51-2\left|\cot\theta_H\right|^{2/3}}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{1/2}}+\frac{1}{2}\overline{K}_2\frac{3+\left|\cot\theta_H\right|^{2/3}}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{3/2}}\right]\exp\left(-W_0\right). \quad (37a)$$

where the WKB exponent is

$$W_0=\frac{2^{17/4}\times 3^{1/4}}{5}S\epsilon^{5/4}\left|\cot\theta_H\right|^{1/6}\left[1+\frac{\epsilon}{2}-\frac{9}{2}\overline{K}_2\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{1/2}\right.$$
$$\left.+\frac{1}{4}\overline{K}_2\frac{2+9\left|\cot\theta_H\right|^{2/3}}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{1/2}}-\overline{K}_2\frac{3+\left|\cot\theta_H\right|^{2/3}}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{3/2}}\right]. \quad (37b)$$

The decay rate at a finite temperature T is found to be

$$\Gamma\left(T\right)=2\operatorname{Im}E_0\left(1-e^{-\hbar\Omega_2\beta}\right)\exp\left(q_2e^{-\hbar\Omega_2\beta}\right). \quad (38)$$

In Fig. 3 we plot the temperature dependence of the tunneling rate for the typical values of parameters for nanometer-scale single-domain ferromagnets: $S = 6000$, $\epsilon = 1 - \overline{H}/\overline{H}_c = 0.01$, $\overline{K}_2 = 0.01$, and $\theta_H = 3\pi/4$. From Fig. 3 one can easily see the crossover from purely quantum tunneling to thermally assisted quantum tunneling. The temperature $T_0^{(0)}$ characterizing the crossover from quantum to thermal regimes can be estimated as $k_B T_0^{(0)} = \Delta U / W_0$, where ΔU is the barrier height, and W_0 is the WKB exponent of the ground-state tunneling. It can be shown that in the cubic potential $(q^2 - q^3)$, the usual second-order phase transition from the thermal to the quantum regimes occurs as the temperature is lowered. The second-order phase transition temperature is given by $k_B T_0^{(2)} = \frac{\hbar\omega_b}{2\pi}$, where $\omega_b = \sqrt{\frac{1}{m}\left|U''(x_b)\right|}$ is the frequency of small oscillations near the bottom of the inverted potential $-U(x)$, and x_b corresponds to the bottom of the inverted potential. For the present case, it is easy to obtain that

$$k_B T_0^{(2)} = \frac{2^{1/4} \times 3^{1/4}}{\pi} (K_1 V) S^{-1} \epsilon^{1/4} \frac{\left|\cot\theta_H\right|^{1/6}}{1 + \left|\cot\theta_H\right|^{2/3}} \left[1 - \frac{\epsilon}{2} + \frac{9}{2}\overline{K}_2 \left(1 + \left|\cot\theta_H\right|^{2/3}\right)^{1/2} \right.$$
$$\left. - \frac{1}{4}\overline{K}_2 \frac{21 - 2\left|\cot\theta_H\right|^{2/3}}{\left(1 + \left|\cot\theta_H\right|^{2/3}\right)^{1/2}} + \overline{K}_2 \frac{3 + \left|\cot\theta_H\right|^{2/3}}{\left(1 + \left|\cot\theta_H\right|^{2/3}\right)^{3/2}} \right],$$

and $k_B T_0^{(0)} = (5\pi/18)\, k_B T_0^{(2)} \approx 0.87 k_B T_0^{(2)}$. For a nanometer-scale single-domain FM particle, the typical values of parameters for the magnetic anisotropy coefficients are $K_1 = 10^8$ erg/cm^3, and $K_2 = 10^5$ erg/cm^3. The radius of the FM particle is about 12 nm and the sublattice spin is 10^6. In Fig. 4, we plot the θ_H dependence of the crossover temperature T_c for typical values of parameters for nanometer-scale ferromagnets at $\epsilon = 0.001$ in a wide range of angles $\pi/2 < \theta_H < \pi$. Fig. 4 shows that the maximal value of T_c is about 0.26K at $\theta_H = 1.76$. The maximal value of T_c as well as Γ is expected to be observed in experiment. If $\epsilon = 0.001$, we obtain that $T_c(135^\circ) \backsim 0.23$K corresponding to the crossover from quantum to classical regime. Note that, even for ϵ as small as 10^{-3}, the angle corresponding to an appreciable change of the orientation of the magnetization vector by quantum tunneling is $\delta_2 = \sqrt{6\epsilon}$ rad$> 4^\circ$. It is quite large enough to distinguish easily between the two states for experimental tests.

IV. MQC AND MQT FOR TETRAGONAL CRYSTAL SYMMETRY

In this section, we study the FM particles with tetragonal crystal symmetry in a magnetic field at arbitrarily directed angles in the ZX plane, which has the magnetocrystalline anisotropy energy

$$E\left(\theta,\phi\right)=K_1\sin^2\theta+K_2\sin^4\theta-K_2'\sin^4\theta\cos\left(4\phi\right)-M_0H_x\sin\theta\cos\phi-M_0H_z\cos\theta+E_0, \tag{39}$$

where K_1, K_2 and K_2' are the magnetic anisotropy coefficients, and $K_1>0$. In the absence of magnetic field, the easy axes of this system are $\pm\widehat{z}$ for $K_1>0$. And the field is applied in the ZX plane as in the previous section. By using the dimensionless parameters defined in Eq. (8), and choosing $K_2'>0$, we find that $\phi=0$ is an easy plane for this system, at which Eq. (38) reduces to

$$\overline{E}\left(\theta,\phi=0\right)=\frac{1}{2}\sin^2\theta+\left(\overline{K}_2-\overline{K}_2'\right)\sin^4\theta-\overline{H}\cos\left(\theta-\theta_H\right)+\overline{E}_0, \tag{40}$$

where $\overline{K}_2'=K_2'/2K_1$. Assuming that $\left|\overline{K}_2-\overline{K}_2'\right|\ll 1$, we obtain the critical magnetic field and the critical angle as

$$\begin{aligned}\overline{H}_c&=\frac{1}{\left[\left(\sin\theta_H\right)^{2/3}+\left|\cos\theta_H\right|^{2/3}\right]^{3/2}}\left[1+\frac{4\left(\overline{K}_2-\overline{K}_2'\right)}{1+\left|\cot\theta_H\right|^{2/3}}\right],\\ \sin\theta_c&=\frac{1}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{1/2}}\left[1+\frac{8}{3}\left(\overline{K}_2-\overline{K}_2'\right)\frac{\left|\cot\theta_H\right|^{2/3}}{1+\left|\cot\theta_H\right|^{2/3}}\right].\end{aligned} \tag{41}$$

Introducing $\delta\equiv\theta-\theta_0$ ($|\delta|\ll 1$ in the small ϵ limit), we derive the energy $\overline{E}\left(\theta,\phi\right)$ as

$$\overline{E}\left(\delta,\phi\right)=\overline{K}_2'\left[1-\cos\left(4\phi\right)\right]\sin^4\left(\theta_0+\delta\right)+\overline{H}_x\left(1-\cos\phi\right)\sin\left(\theta_0+\delta\right)+\overline{E}_1\left(\delta\right), \tag{42}$$

where $\overline{E}_1\left(\delta\right)$ is a function of only δ given by

$$\begin{aligned}\overline{E}_1\left(\delta\right)\ =&\left[\frac{1}{2}\overline{H}_c\sin\left(\theta_c-\theta_H\right)+\left(\overline{K}_2-\overline{K}_2'\right)\sin\left(4\theta_c\right)\right]\left(\delta^3-3\delta^2\eta\right)\\ &+\left[\frac{1}{8}\overline{H}_c\cos\left(\theta_c-\theta_H\right)+\left(\overline{K}_2-\overline{K}_2'\right)\cos\left(4\theta_c\right)\right]\left(\delta^4-4\delta^3\eta+6\delta^2\eta^2-4\delta^2\epsilon\right)\\ &+4\left(\overline{K}_2-\overline{K}_2'\right)\epsilon\delta^2\cos\left(4\theta_c\right).\end{aligned} \tag{43}$$

A. $\theta_H = \pi/2$

For this case, we obtain that $\eta \approx \sqrt{2\epsilon}\left[1 - 4\left(\overline{K}_2 - \overline{K}_2'\right)\right]$, and $\theta_c = \pi/2$. Performing the Gaussian integration over ϕ, we can map the spin system onto a problem of particle with effective mass m moving in the one-dimensional potential well $U(\delta)$. For this case,

$$m = \frac{\hbar S^2}{2VK_1\left[1 - \epsilon + 4\left(\overline{K}_2 - \overline{K}_2'\right) + 16\overline{K}_2'\right]},$$

and

$$U(\delta) = \frac{K_1 V}{4\hbar}\left[1 + 12\left(\overline{K}_2 - \overline{K}_2'\right)\right]\delta^2(\delta - 2\eta)^2. \tag{44}$$

By applying the method similar to that in Sec. III. A, we obtain the low-lying tunnel splitting at degenerate excited levels as $\hbar\Delta E_n = \frac{q_3^n}{n!}(\hbar\Delta E_0)$, where $q_3 = \frac{2^{9/2}S\epsilon^{3/2}}{1-\frac{\epsilon}{2}+8\left(\overline{K}_2-\overline{K}_2'\right)+8\overline{K}_2'}$. The energy shift of the ground state is

$$\hbar\Delta E_0 = \frac{2^{13/4}}{\sqrt{\pi}}(K_1 V)\epsilon^{5/4}S^{-1/2}\left(1 - \frac{\epsilon}{4} + 4\overline{K}_2\right)\exp(-W_0). \tag{45a}$$

where the WKB exponent is

$$W_0 = \frac{2^{5/2}}{3}S\epsilon^{3/2}\left[1 + \frac{\epsilon}{2} - 8\left(\overline{K}_2 - \overline{K}_2'\right) - 8\overline{K}_2'\right]. \tag{45b}$$

Eqs. (45a) and (45b) agree well with the result obtained with the help of the vacuum instanton solution.[5] And the final result of the decay rate at a finite temperature T is $\Gamma(T) = (\Delta E_0)^2\left[\pi\left(1 - e^{-\hbar\Omega_3\beta}\right)/2\Omega_3\right] I_0\left(2q_3 e^{-\hbar\Omega_3\beta/2}\right)$, where $I_0(x)$ is the modified Bessel function.

B. $\pi/2 < \theta_H < \pi$

For $\pi/2 < \theta_H < \pi$, $\eta \approx \sqrt{2\epsilon/3}$, the effective mass m is

$$m = \frac{\hbar S^2\left(1 + |\cot\theta_H|^{2/3}\right)}{2K_1 V\left[1 - \epsilon + 16\overline{K}_2' + \frac{4}{3}\left(\overline{K}_2 - \overline{K}_2'\right)\frac{3-2|\cot\theta_H|^{2/3}}{1+|\cot\theta_H|^{2/3}}\right]}, \tag{46a}$$

and the effective potential is $U(\delta) = 3U_0 q^2 \left(1 - \frac{2}{3}q\right)$, with $q = 3\delta/2\sqrt{6\epsilon}$, and

$$U_0 = \frac{2^{7/4}}{3^{3/2}} \frac{K_1 V}{\hbar} \epsilon^{3/2} \frac{\left|\cot\theta_H\right|^{1/3}}{1 + \left|\cot\theta_H\right|^{2/3}} \left[1 + \frac{4}{3}\left(\overline{K}_2 - \overline{K}_2'\right) \frac{7 - 4\left|\cot\theta_H\right|^{2/3}}{1 + \left|\cot\theta_H\right|^{2/3}}\right]. \tag{46b}$$

For this case, the imaginary part of the low-lying metastable excited levels is $\hbar \operatorname{Im} E_n = \frac{q_4^n}{n!}\left(\hbar \operatorname{Im} E_0\right)$, where $q_4 = \frac{2^{25/4} \times 3^{5/4} S \epsilon^{5/4} \left|\cot\theta_H\right|^{1/6}}{1 - \frac{\epsilon}{2} + 8\overline{K}_2' + \frac{4}{3}\left(\overline{K}_2 - \overline{K}_2'\right) \frac{\left|\cot\theta_H\right|^{2/3} - 2}{1 + \left|\cot\theta_H\right|^{2/3}}}$. The imaginary part of the metastable ground-state level is

$$\begin{aligned} \hbar \operatorname{Im} E_0 = & \frac{3^{13/9} \times 2^{31/8}}{\sqrt{\pi}} \left(K_1 V\right) \epsilon^{7/8} S^{-1/2} \frac{\left|\cot\theta_H\right|^{1/4}}{1 + \left|\cot\theta_H\right|^{2/3}} \\ & \times \left[1 - \frac{\epsilon}{4} + 4\overline{K}_2' + \frac{2}{3}\left(\overline{K}_2 - \overline{K}_2'\right) \frac{12\left|\cot\theta_H\right|^{2/3} - 7}{1 + \left|\cot\theta_H\right|^{2/3}}\right] \exp\left(-W_0\right). \end{aligned} \tag{47a}$$

where

$$W_0 = \frac{2^{17/4} \times 3^{1/4}}{5} S \epsilon^{5/4} \left|\cot\theta_H\right|^{1/6} \left[1 + \frac{\epsilon}{2} - 8\overline{K}_2' - \frac{4}{3}\left(\overline{K}_2 - \overline{K}_2'\right) \frac{\left|\cot\theta_H\right|^{2/3} - 2}{1 + \left|\cot\theta_H\right|^{2/3}}\right]. \tag{47b}$$

The final result of the decay rate at a finite temperature T is $\Gamma(T) = 2 \operatorname{Im} E_0 \left(1 - e^{-\hbar\Omega_4\beta}\right) \exp\left(q_4 e^{-\hbar\Omega_4\beta}\right)$. And the second-order phase transition temperature characterizing the crossover from quantum to thermal regimes is found to be

$$\begin{aligned} k_B T_0^{(2)} = & \frac{2^{1/4} \times 3^{1/4}}{\pi} \left(K_1 V\right) S^{-1} \epsilon^{1/4} \frac{\left|\cot\theta_H\right|^{1/6}}{1 + \left|\cot\theta_H\right|^{2/3}} \\ & \times \left[1 - \frac{\epsilon}{2} + 8\overline{K}_2' + \frac{4}{3}\left(\overline{K}_2 - \overline{K}_2'\right) \frac{5 - 3\left|\cot\theta_H\right|^{2/3}}{1 + \left|\cot\theta_H\right|^{2/3}}\right]. \end{aligned}$$

V. MQC AND MQT FOR HEXAGONAL CRYSTAL SYMMETRY

In this section, we study the hexagonal spin system whose magnetocrystalline anisotropy energy $E_a(\theta, \phi)$ at zero magnetic field can be written as

$$E_a(\theta, \phi) = K_1 \sin^2\theta + K_2 \sin^4\theta + K_3 \sin^6\theta - K_3' \sin^6\theta \cos(6\phi), \tag{48}$$

where K_1, K_2, K_3, and K_3' are the magnetic anisotropic coefficients. The easy axes are $\pm\widehat{z}$ for $K_1 > 0$. When we apply an external magnetic field at an arbitrarily directed angle in the ZX plane, the total energy of this system is given by

$$E\left(\theta,\phi\right)=E_a\left(\theta,\phi\right)-M_0H_x\sin\theta\cos\phi-M_0H_z\cos\theta+E_0, \tag{49}$$

By choosing $K_3'>0$, we take $\phi=0$ to be the easy plane, at which the potential energy can be written in terms of the dimensionless parameters as

$$\overline{E}\left(\theta,\phi=0\right)=\frac{1}{2}\sin^2\theta+\overline{K}_2\sin^4\theta+\left(\overline{K}_3-\overline{K}_3'\right)\sin^6\theta-\overline{H}\cos\left(\theta-\theta_H\right)+\overline{E}_0, \tag{50}$$

where $\overline{K}_3=K_3/2K_1$ and $\overline{K}_3'=K_3'/2K_1$.

Under the assumption that $\left|\overline{K}_2\right|$, $\left|\overline{K}_3-\overline{K}_3'\right|\ll 1$, we obtain the dimensionless critical field $\overline{H}_c$ and the critical angle θ_c as

$$\overline{H}_c=\frac{1}{\left[\left(\sin\theta_H\right)^{2/3}+\left|\cos\theta_H\right|^{2/3}\right]^{3/2}}\left[1+\frac{4\overline{K}_2}{1+\left|\cot\theta_H\right|^{2/3}}+\frac{6\left(\overline{K}_3-\overline{K}_3'\right)}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^2}\right],$$

$$\sin\theta_c=\frac{1}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^{1/2}}\left[1+\frac{8}{3}\overline{K}_2\frac{\left|\cot\theta_H\right|^{2/3}}{1+\left|\cot\theta_H\right|^{2/3}}+8\left(\overline{K}_3-\overline{K}_3'\right)\frac{\left|\cot\theta_H\right|^{2/3}}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^2}\right]. \tag{51}$$

By introducing a small variable $\delta\equiv\theta-\theta_0$ ($|\delta|\ll 1$ in the limit of $\epsilon\ll 1$), the total energy becomes

$$\overline{E}\left(\delta,\phi\right)=\overline{K}_3'\left[1-\cos\left(6\phi\right)\right]\sin^6\left(\theta_0+\delta\right)+\overline{H}_x\left(1-\cos\phi\right)\sin\left(\theta_0+\delta\right)+\overline{E}_1\left(\delta\right), \tag{52}$$

where $\overline{E}_1\left(\delta\right)$ is a function of only δ given by

$$\begin{aligned}\overline{E}_1\left(\delta\right) =& \left[\frac{1}{2}\overline{H}_c\sin\left(\theta_c-\theta_H\right)+\overline{K}_2\sin\left(4\theta_c\right)+4\left(\overline{K}_3-\overline{K}_3'\right)\left(5\sin^3\theta_c\cos^3\theta_c-3\sin^5\theta_c\cos\theta_c\right)\right]\\
&\times\left(\delta^3-3\delta^2\eta\right)+\left[\frac{1}{8}\overline{H}_c\cos\left(\theta_c-\theta_H\right)+\overline{K}_2\cos\left(4\theta_c\right)+3\left(\overline{K}_3-\overline{K}_3'\right)\sin^2\theta_c\left(\sin^4\theta_c\right.\right.\\
&\left.\left.-10\sin^2\theta_c\cos^2\theta_c+5\cos^4\theta_c\right)\right]\left(\delta^4-4\delta^3\eta+6\delta^2\eta^2-4\delta^2\epsilon\right)+\epsilon\delta^2\left[4\overline{K}_2\cos\left(4\theta_c\right)\right.\\
&\left.+12\left(\overline{K}_3-\overline{K}_3'\right)\sin^2\theta_c\left(\sin^4\theta_c-10\sin^2\theta_c\cos^2\theta_c+5\cos^4\theta_c\right)\right].\end{aligned} \tag{53}$$

A. $\theta_H=\pi/2$

For $\theta_H=\pi/2$, i.e., the external magnetic field is applied perpendicular to the anisotropy axis, we obtain that $\theta_c=\pi/2$ and $\eta=\sqrt{2\epsilon}\left[1-4\overline{K}_2-12\left(\overline{K}_3-\overline{K}_3'\right)\right]$. The spin system

can be mapped onto a particle with effective mass m moving in the one-dimensional potential well $U(\delta)$, where

$$m = \frac{\hbar S^2}{2VK_1\left[1-\epsilon-4\overline{K}_2-6\left(\overline{K}_3-\overline{K}_3'\right)-36\overline{K}_3'\right]}, \tag{54a}$$

and

$$U(\delta) = \frac{K_1 V}{4\hbar}\left[1+12\overline{K}_2+30\left(\overline{K}_3-\overline{K}_3'\right)\right]\delta^2(\delta-2\eta)^2. \tag{54b}$$

By applying the similar method, we obtain that the energy shift of the n-th excited level is $\hbar\Delta E_n = \frac{q_5^n}{n!}(\hbar\Delta E_0)$,where

$$q_5 = \frac{2^{9/2}S\epsilon^{3/2}}{1-\frac{\epsilon}{2}+8\overline{K}_2+24\left(\overline{K}_3-\overline{K}_3'\right)+18\overline{K}_3'}.$$

The energy shift of the ground state is

$$\hbar\Delta E_0 = \frac{2^{13/4}}{\sqrt{\pi}}(K_1V)\,\epsilon^{5/4}S^{-1/2}\left[1-\frac{\epsilon}{4}-6\left(\overline{K}_3-\overline{K}_3'\right)+9\overline{K}_3'\right]\exp(-W_0), \tag{55a}$$

and the WKB exponent is

$$W_0 = \frac{2^{5/2}}{3}S\epsilon^{3/2}\left[1+\frac{\epsilon}{2}-8\overline{K}_2-24\left(\overline{K}_3-\overline{K}_3'\right)-18\overline{K}_3'\right]. \tag{55b}$$

The decay rate at a finite temperature T is

$$\Gamma(T) = (\Delta E_0)^2\left[\pi\left(1-e^{-\hbar\Omega_5\beta}\right)/2\Omega_5\right]I_0\left(2q_5e^{-\hbar\Omega_5\beta/2}\right),$$

where

$$\Omega_5 = 2^{3/2}\frac{K_1V}{\hbar S}\epsilon^{3/2}\left[1-\frac{\epsilon}{2}+4\overline{K}_2+6\left(\overline{K}_3-\overline{K}_3'\right)+18\overline{K}_3'\right].$$

B. $\pi/2 < \theta_H < \pi$

For this case, the effective mass m and the effective potential $U(\delta)$ are

$$m = \frac{\hbar S^2\left(1+|\cot\theta_H|^{2/3}\right)}{2K_1V\left[1-\epsilon+\frac{4}{3}\overline{K}_2\frac{3-2|\cot\theta_H|^{2/3}}{1+|\cot\theta_H|^{2/3}}+2\left(\overline{K}_3-\overline{K}_3'\right)\frac{3-4|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^2}+36\overline{K}_3'\frac{1}{1+|\cot\theta_H|^{2/3}}\right]},$$

and

$$U(\delta) = \frac{K_1 V}{\hbar} \frac{|\cot\theta_H|^{1/3}}{1+|\cot\theta_H|^{2/3}} \left[1 + \frac{4}{3}\overline{K}_2 \frac{7-4|\cot\theta_H|^{2/3}}{1+|\cot\theta_H|^{2/3}} + 2\left(\overline{K}_3 - \overline{K}_3'\right) \frac{11-16|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^2} \right]$$
$$\times \delta^2 \left(\sqrt{6\epsilon} - \delta\right).$$

The imaginary part of the metastable excited levels is $\hbar\,\mathrm{Im}\,E_n = \frac{q_6^n}{n!}\left(\hbar\,\mathrm{Im}\,E_0\right)$, and the imaginary part of the ground state is

$$\hbar\,\mathrm{Im}\,E_0 = \frac{3^{7/9}\times 2^{31/8}}{\sqrt{\pi}} (K_1 V)\,\epsilon^{7/8} S^{-1/2} \frac{|\cot\theta_H|^{1/4}}{1+|\cot\theta_H|^{2/3}} \left[1 - \frac{\epsilon}{4} + \frac{2}{3}\overline{K}_2 \frac{12|\cot\theta_H|^{2/3}-7}{1+|\cot\theta_H|^{2/3}} \right.$$
$$\left. 2\left(\overline{K}_3 - \overline{K}_3'\right) \frac{9-13|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^2} + 9\overline{K}_3' \frac{1}{1+|\cot\theta_H|^{2/3}} \right] \exp(-W_0), \tag{56a}$$

where the WKB exponent is

$$W_0 = \frac{2^{17/4}\times 3^{1/4}}{5} S\epsilon^{5/4} |\cot\theta_H|^{1/6} \left[1 - \frac{\epsilon}{4} + \frac{4}{3}\overline{K}_2 \frac{2-|\cot\theta_H|^{2/3}}{1+|\cot\theta_H|^{2/3}} \right.$$
$$\left. +4\left(\overline{K}_3 - \overline{K}_3'\right) \frac{2-3|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^2} - 18\overline{K}_3' \frac{1}{1+|\cot\theta_H|^{2/3}} \right], \tag{56b}$$

and

$$q_6 = \frac{2^{25/4}\times 3^{5/4} S\epsilon^{5/4} |\cot\theta_H|^{1/6}}{1 - \frac{\epsilon}{2} - \frac{4}{3}\overline{K}_2 \frac{2-|\cot\theta_H|^{2/3}}{1+|\cot\theta_H|^{2/3}} - 4\left(\overline{K}_3 - \overline{K}_3'\right) \frac{2-3|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^2} + 18\overline{K}_3' \frac{1}{1+|\cot\theta_H|^{2/3}}}. \tag{56c}$$

The final result of the decay rate at a finite temperature T is $\Gamma(T) = 2\,\mathrm{Im}\,E_0 \left(1 - e^{-\hbar\Omega_6\beta}\right) \exp\left(q_6 e^{-\hbar\Omega_6\beta}\right)$, where

$$\Omega_6 = 2^{5/4}\times 3^{1/4} \frac{K_1 V}{\hbar S} \epsilon^{1/4} \frac{|\cot\theta_H|^{1/6}}{1+|\cot\theta_H|^{2/3}} \left[1 - \frac{\epsilon}{2} + \frac{4}{3}\overline{K}_2 \frac{5-3|\cot\theta_H|^{2/3}}{1+|\cot\theta_H|^{2/3}} \right.$$
$$\left. +2\left(\overline{K}_3 - \overline{K}_3'\right) \frac{7-10|\cot\theta_H|^{2/3}}{\left(1+|\cot\theta_H|^{2/3}\right)^2} + 18\overline{K}_3' \frac{1}{1+|\cot\theta_H|^{2/3}} \right].$$

The second-order phase transition temperature characterizing the crossover from quantum to thermal regimes is found to be

$$k_B T_0^{(2)} = \frac{2^{1/4} \times 3^{1/4}}{\pi} \left(K_1 V\right) S^{-1} \epsilon^{1/4} \frac{\left|\cot\theta_H\right|^{1/6}}{1+\left|\cot\theta_H\right|^{2/3}} \left[1 - \frac{\epsilon}{2} + \frac{4}{3}\overline{K}_2 \frac{5 - 3\left|\cot\theta_H\right|^{2/3}}{1+\left|\cot\theta_H\right|^{2/3}} \right.$$
$$\left. +2\left(\overline{K}_3 - \overline{K}_3'\right) \frac{7 - 10\left|\cot\theta_H\right|^{2/3}}{\left(1+\left|\cot\theta_H\right|^{2/3}\right)^2} + 18\overline{K}_3' \frac{1}{1+\left|\cot\theta_H\right|^{2/3}} \right].$$

VI. CONCLUSIONS

In summary, we have theoretically investigated the quantum tunneling of the magnetization vector between excited levels in single-domain FM nanoparticles in the presence of an external magnetic field at arbitrary angle. We consider the FM particles with the general structure of magnetocrystalline anisotropy. By calculating the nonvacuum instanton in the spin-coherent-state path-integral representation, we obtain the analytic formulas for the tunnel splitting between degenerate excited levels and the imaginary parts of the metastable excited levels in the low barrier limit for the external magnetic field perpendicular to the easy axis ($\theta_H = \pi/2$), and for the field at an angle between the easy and hard axes ($\pi/2 < \theta_H < \pi$). The temperature dependences of the decay rates are clearly shown for each case. The low-energy limit of our results agrees well with that of ground-state spin tunneling. One important conclusion is that the tunneling rate and the tunnel splitting at excited levels depend on the orientation of the external magnetic field distinctly. Even a small misalignment of the field with $\theta_H = \pi/2$ orientation can completely change the results of the tunneling rates. Another interesting conclusion concerns the field strength dependence of the WKB exponent in the tunnel splitting or the tunneling rate. It is found that in a wide range of angles, the $\epsilon \left(= 1 - \overline{H}/\overline{H}_c\right)$ dependence of the WKB exponent is given by $\epsilon^{5/4}$ (see Eq. (37b)), not $\epsilon^{3/2}$ for $\theta_H = \pi/2$ (see Eq. (21b)). As a result, we conclude that both the orientation and the strength of the external magnetic field are the controllable parameters for the experimental test of the phenomena of quantum tunneling and coherence of the magnetization vector between excited levels in single-domain FM nanoparticles at sufficiently low temperatures. If the experiment is to be performed, there are three control

parameters for comparison with theory: the angle of the external magnetic field θ_H, the strength of the field in terms of ϵ, and the temperature T. Furthermore, the θ_H dependence of the crossover temperature T_c and the angle corresponding to the maximal value of T_c are expected to be observed in further experiments.

In order to avoid the complications due to distributions of particle size and shape, some groups have tried to study the temperature and field dependence of magnetization reversal of individual magnets. Recently, Wernsdorfer and co-workers have performed the switching field measurements on individual ferrimagnetic and insulating BaFeCoTiO nanoparticles containing about 10^5-10^6 spins at very low temperatures (0.1-6K).[8] They found that above 0.4K, the magnetization reversal of these particles is unambiguously described by the Néel-Brown theory of thermal activated rotation of the particle's moment over a well defined anisotropy energy barrier. Below 0.4K, strong deviations from this model are evidenced which are quantitatively in agreement with the predictions of the MQT theory without dissipation.[4] The BaFeCoTiO nanoparticles have a strong uniaxial magnetocrystalline anisotropy.[8] However, the theoretical results presented here may be useful for checking the general theory in a wide range of systems, with more general magnetic anisotropy. The experimental procedures on single-domain FM nanoparticles of Barium ferrite with uniaxial symmetry[8] may be applied to the systems with more general symmetries. Note that the inverse of the WKB exponent B^{-1} is the magnetic viscosity S at the quantum-tunneling-dominated regime $T \ll T_c$ studied by magnetic relaxation measurements.[1] Therefore, the quantum tunneling of the magnetization should be checked at any θ_H by magnetic relaxation measurements. Over the past years a lot of experimental and theoretical works were performed on the spin tunneling in molecular Mn_{12}-Ac[23] and Fe_8[24,20] clusters having a collective spin state $S = 10$ (in this paper $S = 10^3 - 10^5$). These measurements on molecular clusters with $S = 10$ suggest that quantum phenomena might be observed at larger system sizes with $S \gg 1$. Further experiments should focus on the level quantization of collective spin states of $S = 10^2$-10^4.

The theoretical calculations performed in this paper can be extended to the AFM parti-

cles, where the relevant quantity is the excess spin due to the small noncompensation of two sublattices. Work along this line is still in progress. We hope that the theoretical results presented in this paper may stimulate more experiments whose aim is observing quantum tunneling and quantum coherence in nanometer-scale ferromagnets.

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REFERENCES

[1] For a review, see *Quantum Tunneling of Magnetization*, edited by L. Gunther and B. Barbara (Kluwer, Dordrecht, 1995); and E. M. Chudnovsky and J. Tejada, *Macroscopic Quantum Tunneling of the Magnetic Moment* (Cambridge University Press, 1997).

[2] I. Ya Korenblit and E. F. Shender, Zh. Eksp. Teor. Fiz. **75**, 1862 (1978)[Sov. Phys. JETP **48**, 937 (1978)].

[3] O. B. Zaslavskii, Phys. Rev. B **42**, 992 (1990).

[4] M. -G. Miguel and E. M. Chudnovsky, Phys. Rev. B **54**, 388 (1996).

[5] G. -H. Kim and D. S. Hwang, Phys. Rev. B **55**, 8918 (1997).

[6] G. -H. Kim, Phys. Rev. B **57**, 10688 (1998).

[7] D. A. Garanin, X. M. Hidalgo, and E. M. Chudonovsky, Phys. Rev. B **57**, 13639 (1998).

[8] W. Wernsdorfer, E. B. Orozco, K. Hasselbach, A. Benoit, D. Mailly, O. Kubo, H. Nakano, and B. Barbara, Phys. Rev. Lett. **79**, 4014 (1997).

[9] Rong Lü, Jia-Lin Zhu, Xiao-Bing Wang, and Lee Chang, Phys. Rev. B **60**, 4101 (1999).

[10] D. Loss, D. P. DiVicenzo, and G. Grinstein, Phys. Rev. Lett. **69**, 3232 (1992).

[11] J. V. Delft and G. L. Henley, Phys. Rev. Lett. **69**, 3236 (1992).

[12] A. Garg, Europhys. Lett. **22**, 205 (1993).

[13] H. B. Braun and D. Loss, Europhys. Lett. **31**, 555 (1995).

[14] W. Wernsdorfer and R. Sessoli, Science **284**, 133 (1999).

[15] B. Barbara and E. M. Chudnovsky, Phys. Lett. **A145**, 205 (1990); E. M. Chudnovsky, J. Magn. Magn. Mater. **140-144**, 1821 (1995).

[16] L. D. Landau and E. M. Lifshita, *Quantum Mechanics* (Pergamon, London, 1965).

[17] E. B. Bogomolny and V. A. Fateyev, Phys. Lett. B **71**, 93 (1977); see also Ref. 6 and references therein.

[18] A. Garg, Phys. Rev. B **51**, 15161 (1995).

[19] U. Weiss, *Quantum Dissipative Systems,* (World Scientific, 1999).

[20] For a review of recent development on spin tunneling in molecular cluster and the dissipation effects, see I. Tupitsyn and B. Barbara, cond-mat/0002180 and references therein; B. Barbara, L. Thomas, F. Lionti, I. Chiorescu, and A. Sulpice, J. Magn. Magn. Mater. **200**, 167 (1999).

[21] A. Garg and G. -H. Kim, Phys. Rev. Lett. **63**, 2512 (1989); Phys. Rev. B **43**, 712 (1991); H. Simanjuntak, J. Low Temp. Phys. **90**, 405 (1992); P. C. E. Stamp, Phys. Rev. Lett. **66**, 2802 (1991); E. M. Chudnovsky, O. Iglesias, and P. C. E. Stamp, Phys. Rev. B **46**, 5392 (1992); A. Garg, Phys. Rev. Lett. **70**, 1541 (1993); **74**, 1458 (1995); J. Appl. Phys. **76**, 6168 (1994); G. Tatara and H. Fukuyama, Phys. Rev. Lett. **72**, 772 (1994).

[22] E. M. Chudnovsky and D. P. DiVincenzo, Phys. Rev. B **48**, 10548 (1993).

[23] C. Paulsen, J. -G. Park, B. Barbara, R. Sessoli, and A. Caneschi, J. Magn. Magn. Mater. **140-144**, 379 (1995); C. Paulsen and J. -G. Park, in *Quantum Tunneling of Magnetization*, edited by L. Gunther and B. Barbara (Kluwer, Dordrecht, 1995); M. A. Novak and R. Sessoli, in *Quantum Tunneling of Magnetization*, edited by L. Gunther and B. Barbara (Kluwer, Dordrecht, 1995); J. M. Hernández, X. X. Zhang, F. Luis, J. Bartolomé, J. Tejada, and R. Ziolo, Europhys. Lett. **35**, 301 (1996); L. Thomas, F. Lionti, R. Ballou, D. Gatteschi, R. Sessoli, and B. Barbara, Nature (London) **383**, 145 (1996); J. R. Friedman, M. P. Sarachik, J. Tejada, and R. Ziolo, Phys. Rev. Lett. **76**, 3820 (1996); J. M. Hernández, X. X. Zhang, F. Luis, J. Tejada, J. R. Friedman, M. P. Sarachik, and R. Ziolo, Phys. Rev. B **55**, 5858 (1997); F. Lionti, L. Thomas, R. Ballou, B. Barbara, A. Sulpice, R. Sessoli, and D. Gatteschi, J. Appl. Phys. **81**, 4608 (1997); D. A. Garanin and E. M. Chudnovsky,

Phys. Rev. B **56**, 11102 (1997).

[24] A.-L. Barra, P. Debrunner, D. Gatteschi, C. E. Schulz, R. Sessoli, Europhys. Lett. **35**, 133 (1996); C. Sangregorio, T. Ohm, C. Paulsen, R. Sessoli, and D. Gatteschi, Phys. Rev. Lett. **78**, 4645 (1997).

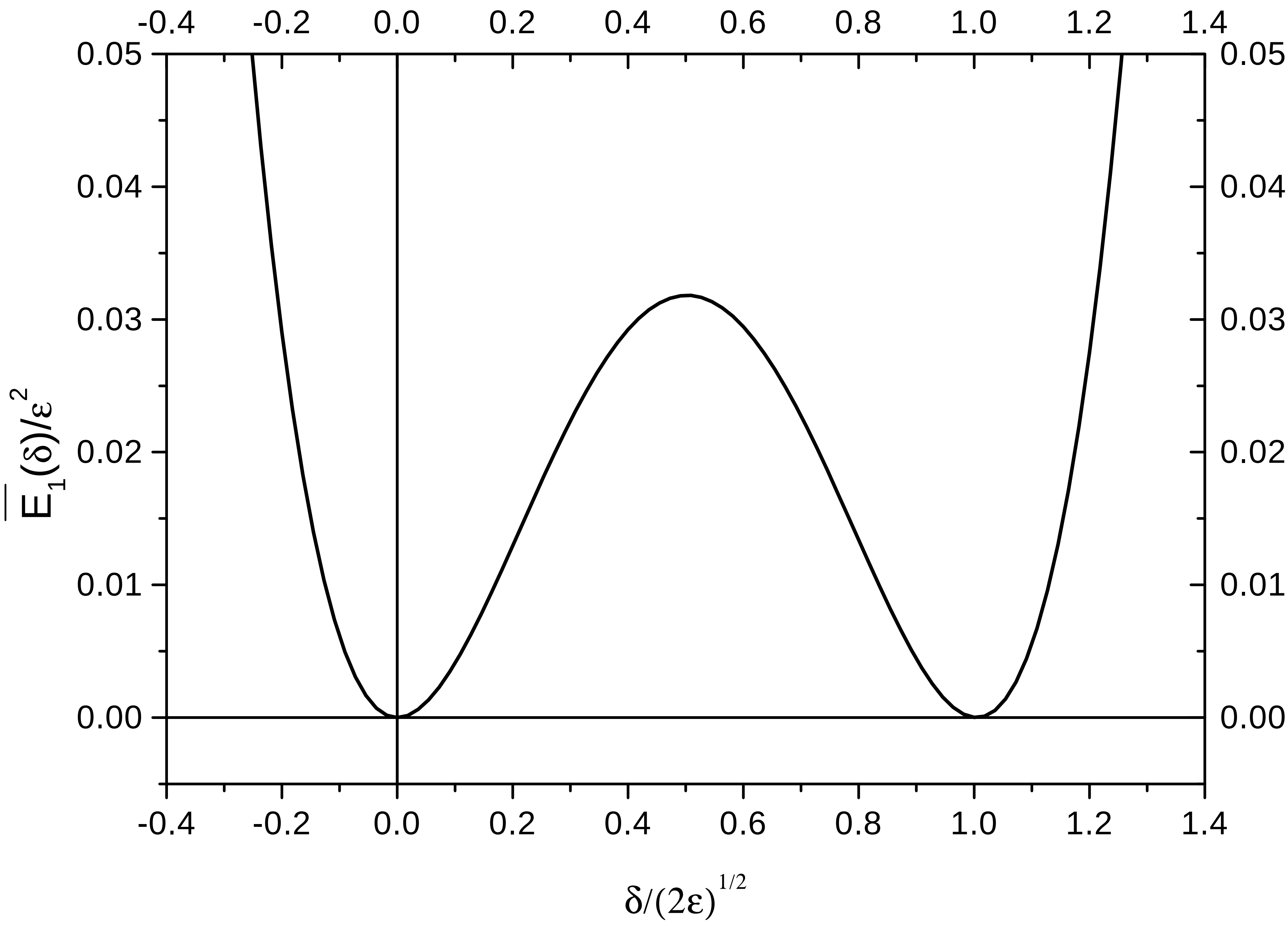
Figure Captions:

Fig. 1 The $\delta\,(=\theta-\theta_0)$ dependence of the effective potential $\overline{E}_1\,(\delta)$ for $\theta_H=\pi/2$ (MQC).

Fig. 2 The $\delta\,(=\theta-\theta_0)$ dependence of the effective potential $\overline{E}_1\,(\delta)$ for $\theta_H=3\pi/4$ (MQT). Here, $\overline{K}_2=0.001$.

Fig. 3 The temperature dependence of the relative decay rate $\Gamma\,(T)\,/\Gamma\,(T=0\mathrm{K})$ for FM particles in a magnetic field with a range of angles $\pi/2<\theta_H<\pi$. Here, $S=6000$, $\epsilon=1-\overline{H}/\overline{H}_c=0.01$, $\overline{K}_2=0.01$, and $\theta_H=3\pi/4$.

Fig. 4 The θ_H dependence of the crossover temperature T_c for $\pi/2<\theta_H<\pi$.



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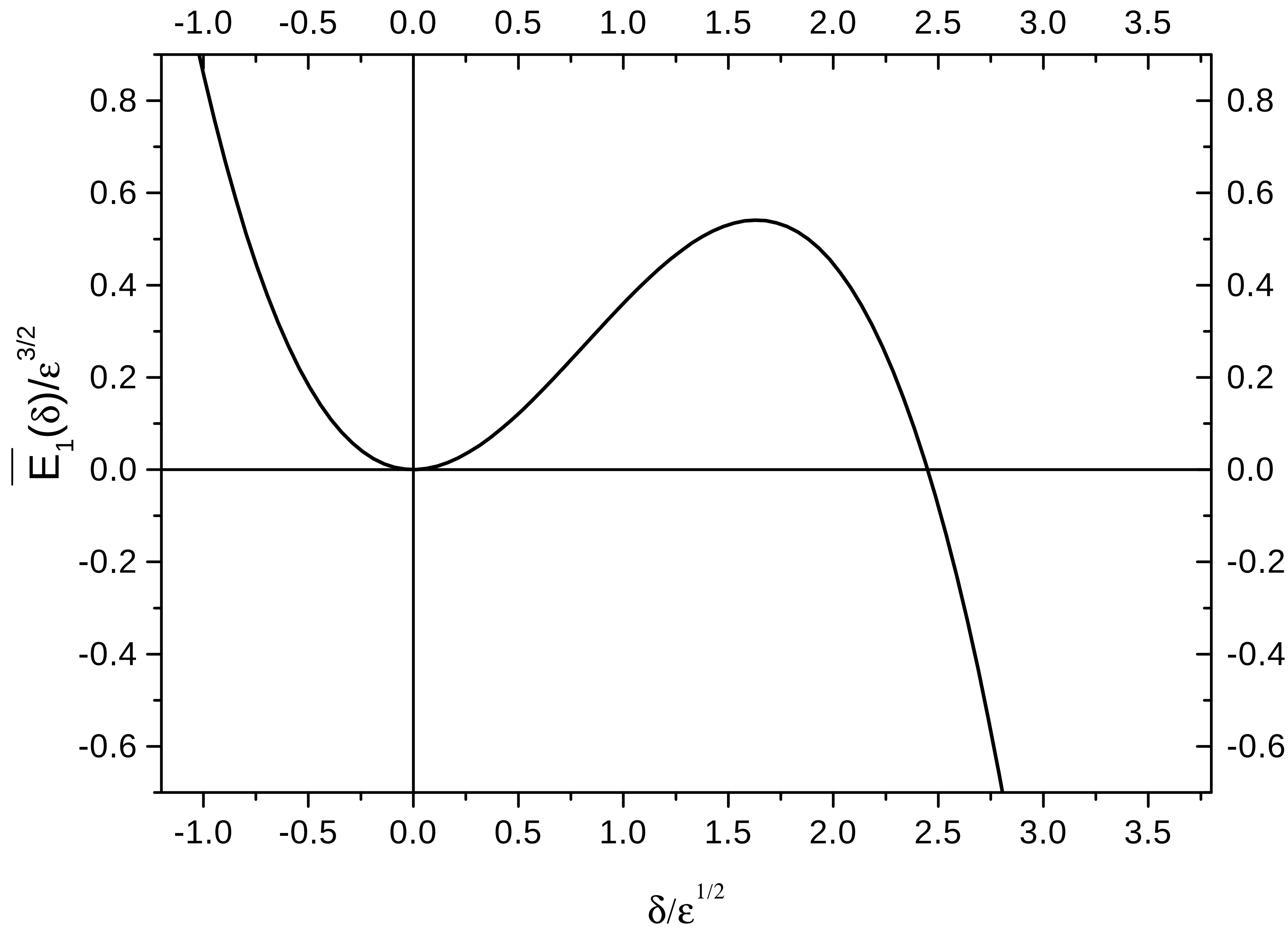
-0.2
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
0.05
0.04
0.03
0.02
0.01
0.00
$\overline{E}_1(\delta)/\varepsilon^2$
$\delta/(2\varepsilon)^{1/2}$



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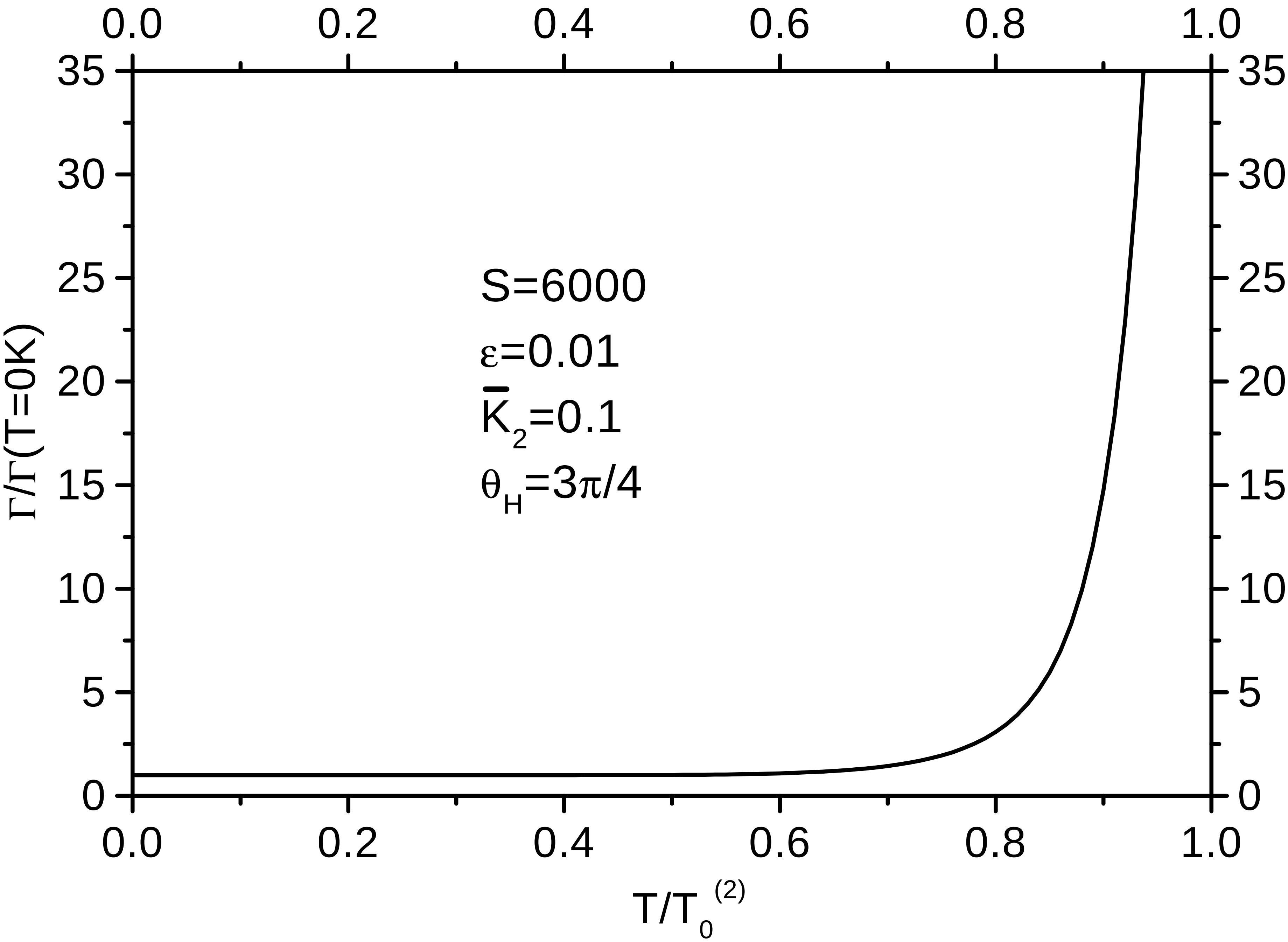
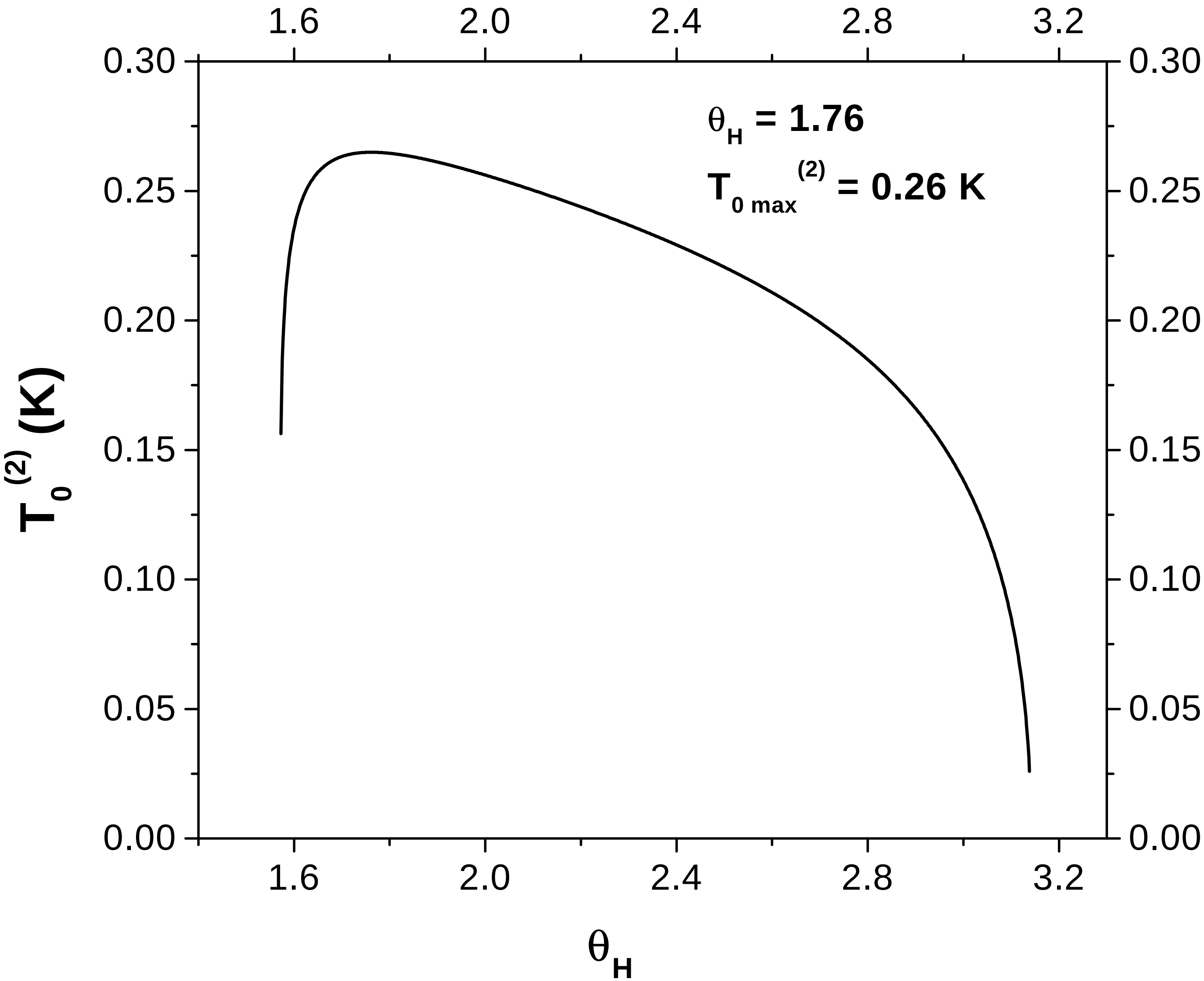
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
$\overline{E_1}(\delta)/\varepsilon^{3/2}$
$\delta/\varepsilon^{1/2}$



S=6000
ε=0.01
$\bar{K}_2$=0.1
θ_H=3π/4
Γ/Γ(T=0K)
$T/T_0^{(2)}$
0.0 0.2 0.4 0.6 0.8 1.0
0 5 10 15 20 25 30 35



1.6
2.0
2.4
2.8
3.2
0.30
0.25
0.20
0.15
0.10
0.05
0.00
θ_H = 1.76
$T_{0\,max}^{(2)}$ = 0.26 K
$T_0^{(2)}$ (K)
θ_H